COMPARISON BETWEEN EXPERIMENTAL AND CALCULATED SPECTRA OF CARBON DIOXIDE AND WATER VAPOR

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ABSTRACT

Line by line calculations of absorption spectra have been carried out for the 2.0 and the 2.7 micron bands of carbon dioxide and for the 2.7 micron band of water vapor. The parameters for these calculations are taken from published listings for these bands. The calculations are compared with experimental data of the same spectral resolution, this being a fraction of a wave number. The comparisons show the line positions to be in good agreement with the experimental data. The 2.0 micron bands of carbon dioxide differ from the calculations by a small amount, therefore, adjustment of parameters is required for the experimental and calculated values to agree in the 2.7 micron bands of carbon dioxide. There are large differences between calculated and experimental spectra in the 2.7 micron band of water vapor throughout much of the spectra.

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1. INTRODUCTION

The analysis of infrared absorption spectra requires only a few parameters for each of the lines involved in the spectra, because each band is composed of so many lines that a very large number of parameters are required. These include the line positions, strengths and half-widths. In the case of atmosphere absorption spectra, the temperature dependence of the line strengths is taken easily into account by the use of the energy of the lower state involved in the transition and well known statistical mechanics relations. The temperature dependence of the half-width, which is of less importance than the line strength, might be described as almost uninvestigated. It has been calculated for water vapor by Benedict and Kaplan[1] and found to be different from the dependence usually assumed on the basis of the Van Vleck-Weisskopf[2] theory of line shapes, but the validity of model upon which the calculations are based has been questioned by Benedict and Herman.[3] Experimental data on this problem is essentially nonexistent.

The determination of the parameters for individual lines is a rather difficult process because of the overlapping of other lines and because of the distortion of the lines due to the finite resolving power of the spectrometer used in observing the lines. The effects of the spectrometer can be removed by integrating the absorption over a region of the spectra, thus providing information about many lines rather than about a single line. Some technique involving the effective loss of spectral resolution, either by using integrated absorption or pressurizing the absorbing gas to increase the line widths and thus decrease the amount of distortion to the true absorption spectra by the finite resolving power of the spectrometer is usually used to determine the strengths of all the lines in a particular absorption region. In the usual case this region can be picked so as to involve lines primarily from only one vibration state. The theory of distribution of lines within a particular transition according to the rotational quantum numbers is then used to determine the strengths of individual lines. The "hot" bands are then assigned a strength on the basis of harmonic oscillator wave functions. This is necessary because their absorption is sufficiently great to require consideration, but too small in relation to other neighboring lines to determine by the techniques mentioned above.

The line positions are the most readily determined of the absorption parameters required. The stronger lines can be directly measured and a power series in terms of the quantum numbers determined to assign

the positions of weak or blended lines. Difficulties may be encountered due to resonance or for the isotopic states, but there are no fundamental problems involved, although the fact that the weaker overlapped states cannot be measured to as high a degree of accuracy as the more prominent states results in a tendency for these states to not be reported.

A tabulation of the lines and their associated parameters has been published for the 2.0 micron and for the 2.7 micron bands[4] of CO_2, and for the 2.7 micron bands of H_2O.[5] Except for some narrow regions of the 2.7 micron water band[6] no comparison between these absorption parameters and experimental data are available. In order to establish the degree of accuracy which can be expected in computed spectra using these parameters, spectra have been calculated at paths and pressures corresponding to available experimental data throughout each of these bands. It should be possible to greatly improve the knowledge of the band parameters by comparisons of this sort, however it would require spectra at several paths and pressures. The difficulty of preparing the experimental data in a form suitable for input to the computer has resulted in the present comparisons not being used to improve the parameters, but it is felt that other workers in the field may need an indication of the accuracy of the parameters, and they are being reported here for that purpose.

2.   THE CO_2 BANDS

In the report of Calfee and Benedict the 2.7 micron and 2.0 micron carbon dioxide bands are identified by their type, i.e., the rotational quantum number l for the upper and lower state, their ground state energy, and an assigned band number. The more usual identification by means of the complete set of lower and upper state quantum numbers can be determined from this information, but inasmuch as this report is a comparison of their results with the experimental results of Burch, the identification system of the report will be followed. However, when a state has more than one number only one of the numbers will be referred to. A computational error has resulted in the sum of the strengths of some bands in the listing being different from the band strength. The line strengths have been computed again to correct this error, and the corrected values used for the present work.

Figure 1 shows a comparison between a calculated and observed spectrum. These correspond to an equivalent pressure of 0.12 atmospheres and a path of 50.33 atmosphere cm of carbon dioxide at a

temperature of 296°K. The experimental spectrum was provided by Dr. Darrell E Burch in advance of its publication.[7] The calculated spectrum has been degraded in resolution by the application of a triangular slit function. The width of this slit is not varied throughout the spectra. It is interesting to notice how closely this triangle is reproduced in the weak lines occurring near 3500 cm^{-1}. This implies that the true slit function of the spectrometer is approximately the shape of the experimental lines in this same region.

The spectra are composed of many bands, mainly due to the Σ-Σ bands, numbers 5 and 15, centered at 3714.76 and 3612.81, respectively. The spectra seem to indicate that the listed band strength of the former is too small and that the latter is too large, however an incorrect value of the half-width for the lines could result in the absorption being incorrect. The value suggested in the listing of the band for the half-width at one atmosphere is 0.08 cm^{-1}, but the values of the integrated absorption shown in Table I for each of the bands at several different pressures and half-widths indicates that 0.07 cm^{-1} is a better value. This is the value used for the calculations. Work which is now being completed indicates that it is the line strengths which are in error rather than the half-widths. The line shape was taken to be purely Lorentzian.

There are several of the weaker bands which can be seen to be in error in their strengths, such as state 26 centered at 3527.71 cm^{-1}. The greatest error seems to be in state 19 centered at 3580.29 which is much too strong. It is largely this state that results in the poor agreement in the interval between 3540 and 3600 cm^{-1}. The apparent disagreement in the line positions in this interval is, at least in a large part, due to these incorrect strengths. The blending of lines results in a shift in the apparent position of the lines, and so if the strength of one of the lines is incorrect the blending will result in a different amount of shift. The isotopic state 10 of $C^{12}O^{16}O^{18}$ centered at 3675.11 should also be stronger than the listed strength. This can be seen in the lines occurring at 3662.41, 3671.22 and 3655.15 cm^{-1}. The line positions in the listing seem to be quite good.

A similar comparison between calculated and observed spectra for the 2.0 micron band is given in Figure 2. These spectra correspond to a path of 330 atmosphere cm at an effective pressure of 1.0 atmosphere.[8] The calculated spectrum has been degraded in resolution by the application of a Gaussian slit function. This seems to more nearly approximate the spectrometer slit function than a triangle. The

wavelength interval is great enough in this spectra to require that the width of the slit function be varied as the inverse of the wavenumber squared so as to keep the spectral resolution of a constant in width in terms of wavelength. This is because the wavelength resolution of a grating spectrometer is constant, while the wavenumber resolution is a function of the wavenumber.

The stronger bands in this region are state 38, centered at 4853.58 cm^{-1}, state 26, centered at 4977.79 and state 19, centered at 5099.62. States 19 and 38 are in good agreement with the experimental data when a half-width of 0.08 cm^{-1} is used, although the calculated absorption of state 26 is considerably less than the experimental.

It can also be seen from the lines around 4780.0 that state 40, centered at 4807.65 is much too weak, which is the cause of the dis-agreement in the lines near 4820 cm^{-1}. This is one of the states in which the listed strengths are much smaller than the band strength would indicate, due to the previously mentioned computation error. The sum of the listed lines in state 40, the state having even parity of J", and state 41, which has odd parity of J", is 14.53 cm/atm cm instead of 19.40 as indicated by the band strength.

The Benedict modification[9] of the Lorentz line shape was used in calculating these spectra. That is, the shape function was taken as

$$k_\nu = \frac{a/\pi}{(\nu - \nu_0)^2 + a^2} \qquad |\nu - \nu_0| \leq 0.5, \qquad (1a)$$

$$k_\nu = \frac{(a/\pi)\exp(-|\nu - \nu_0|^a b)}{(\nu - \nu_0)^2 + a^2} \qquad |\nu - \nu_0| > 0.5. \qquad (1b)$$

The values of a and b were taken to be 0.46 by analogy to the 4.3 micron band of carbon dioxide. There is little doubt that these values are incor-rect. The constants are known to have different values for different bands. The ratio of the pressure of the broadening gas to the carbon dioxide pressure is different than is found in the atmosphere. Still, one would expect better agreement in the wings of the bands at 5010 and 5140 than is indicated. The use of different values of a and b does not result in a great deal of improvement at these wavenumbers, and so it is probable that a different functional form of the Benedict modification should be used for wavenumbers nearer the line center. This should be a function which causes the line shape to decrease faster for regions about 10 cm^{-1} from the line center.

3.   THE 2.7 MICRON WATER VAPOR BAND

The absorption bands of water vapor are much more complex
than those of carbon dioxide. The molecule is of the asymmetric rotor
type which results in very little regularity in the spectra. In addition,
the half-widths of the lines differ from line to line. The agreement
between experimental and theoretical spectra[10] is, as one might expect,
not nearly so good. The variation of absorption in different regions is
large enough that it was felt best to use two different groups of experi-
mental data. One group is at an effective pressure of 0.332 atmosphere
with an optical path of 0.0180 gm/cm^2. This sample, shown in Figure 3,
covers the wavenumber interval from 2950 cm^{-1} to 3460 cm^{-1}. The
other sample is at an effective pressure of 0.199 atmosphere and cor-
responds to an optical path of 0.00169 gm/cm^2. It covers the region
from 3470 cm^{-1} to 4050 cm^{-1} and is illustrated in Figure 4.

The water vapor spectra were calculated on the basis of a pure
Lorentz shaped line. The spectra were then degraded by the application
of a Gaussian slit function whose width varied as the inverse square of
the frequency at the center of the slit function. At first glance the agree-
ment appears to be quite good, mainly due to the fact that the spectra
are plotted in terms of transmittance, which is usually near to unity.
If the fractional error in absorptance is considered the errors seem
much larger.

There is considerably more detail in these spectra than can be
discussed, so no attempt will be made to call attention to each of the
observed differences. The general trend seems to be for the strongest
lines to appear to be fairly accurate, and the greatest disagreement to
be in lines of intermediate strength. Another point of interest is that
the absorption in the regions where only very weak lines occur is usually
greater in the experimental data than in the calculated spectra. This
indicates that either the weak lines are not so weak as the listing shows,
or else the Lorentz line is a very poor choice for the line shape.

The integrated absorption within a band is a function of the upper
and lower wavenumbers of the interval integrated over the equation.

$$A_{\nu', \nu''} = \int_{\nu'}^{\nu''} (1 - T(x))\, dx \qquad (2)$$

If ν' is held constant, then the integrated absorptance as a function of ν'' is a cumulative measure of the integrated absorptance. The integrated absorptance experimentally determined and integrated absorptance calculated for each of the samples is compared in Figure 5 for the interval from 2950 cm^{-1} to 3460 cm^{-1}, and in Figure 6 for the interval from 3470 cm^{-1} to 4050 cm^{-1}.

The experimental absorptance in the lower wavenumber interval is, in Figure 5, constantly greater by about 25%, over an interval of a few wavenumbers. Over the entire interval of 400 wavenumbers the integrated absorptance in the two cases differs by 11 cm^{-1}, or on the average, the absorptance in the calculated spectra is less by 0.0275 when the average absorptance is 0.09. The experimental absorptance continues to be greater than the calculated absorptance in the first part of Figure 6, but after 3670 cm^{-1} this trend reverses and with the exception of a small region near 3760 cm^{-1}, the calculated values show the greater increase.

An attempt was made to improve the agreement between the experimental and the calculated spectra over a small region. It was possible to obtain a great improvement, thus the parameters which could be adjusted so as to obtain the improvement were not unique. The values of the half-width were not altered, although in several cases either shifting the position of a line nearer to a neighboring line, which overlapped it, or decreasing its strength could be used to decrease the absorption by the two lines. The experimental spectra, the spectra calculated by the listed line parameters, and the spectra calculated by the modified set of parameters are shown in Figure 7. The original and the modified line positions and strengths for lines having strengths greater than 1 cm^{-1}/gm cm^{-2} are listed in Table II. It should be stressed that these modified values give the agreement between experimental and calculated spectra shown in Figure 7, although it is not possible to determine from this single case whether the proper parameter was adjusted. Spectra at different pressures should enable one to determine if the adjustments in line positions are correct. It is definite that some of the strengths are off by a factor larger than two. In other regions of the spectra some lines can be found which have strengths in error by almost a factor of ten.

4.   DISCUSSION

The agreement obtained between the experimental and calculated carbon dioxide spectra cannot be considered to be incorrect from the

present state of our knowledge of infrared absorption processes. The strength of the 4977 cm^{-1} band would appear to be about 15% greater than the listed value. Further studies which are still in progress indicate that the strength of the 2.7 micron bands should be reduced by about 75% rather than reducing the half-width. These bands are probably among the best known of those in the infrared region but from the view of using bands such as these for quantitative work it is interesting to note that when the integrated absorptance from 3664 to 3764 cm^{-1} is 70 cm^{-1}, a 10% change in the amount of absorber, only results in 2.5% change in the integrated absorptance. Thus a much better understanding of the bands is still desirable.

The disagreement in the strength of the "hot" states is of considerable importance. Some of these states are in Fermi resonance with other states, and this could cause the band strengths to be altered, however it seems more likely that the determination of band strengths by the use of harmonic oscillator approximations of the wave functions is insufficient. Because of the available time and the difficulty of preparing data for input to the computer from the reports of experimental data no attempt has been made to improve upon the band parameters. It is also understood that a new version of these parameters is now being prepared.

It is easy to see how the technique of refining molecular parameters by direct comparison of experimental and computed spectra enables much more accurate values to be determined than by the presently used methods. The main requirement is that data at several pressures be available for eliminating uncertainty due to the half-width. The knowledge of the shifting of the line due to blending obtained by calculations could also be used for improving the accuracy of line positions of overlapping bands. The use of efficient programs for calculating the spectra makes the calculations practical on present computers. For example, about one minute of time is required on a C.D.C. 6600 Computer for calculating 100 cm^{-1} of spectra when 3000 lines are present in the spectra.

The water vapor calculations are not nearly as good as those of the carbon dioxide. It is difficult to justify the use of the different half-widths for different lines in view of the uncertainty in the line strengths. The adjusted parameters used to obtain the agreement shown in Figure 7 also causes doubts about how the positions of the blended lines are known. In a recently released listing of line parameters for the 1.9

and 6.3 micron bands of water vapor[11] an F factor is used in adjusting the strengths of the lines. It is hoped that such corrections will be applied to the 2.7 micron bands in the future.
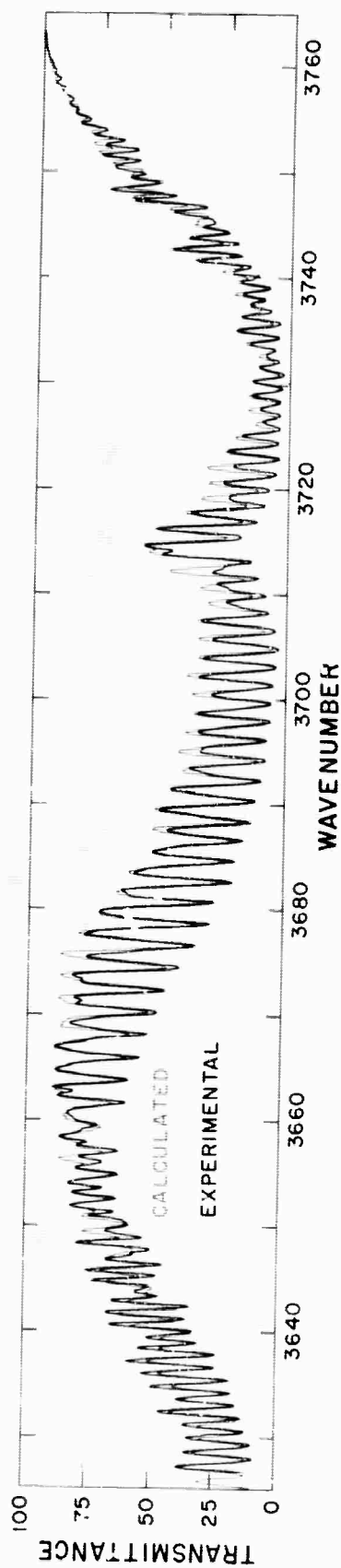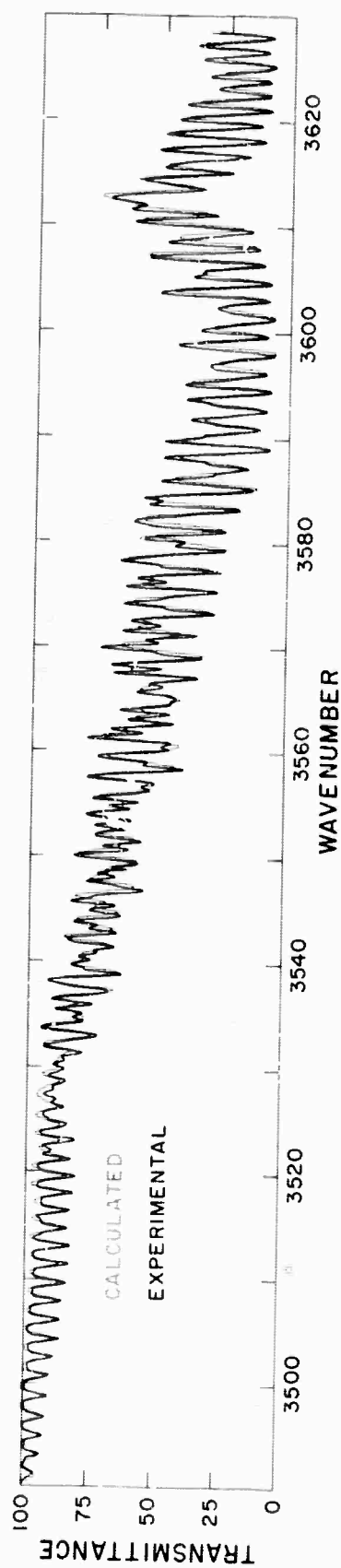
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Figure 1.   The 2.7 micron bands of carbon dioxide at 0.12 atmospheres equivalent pressure and with 50.33 atmosphere cm path.
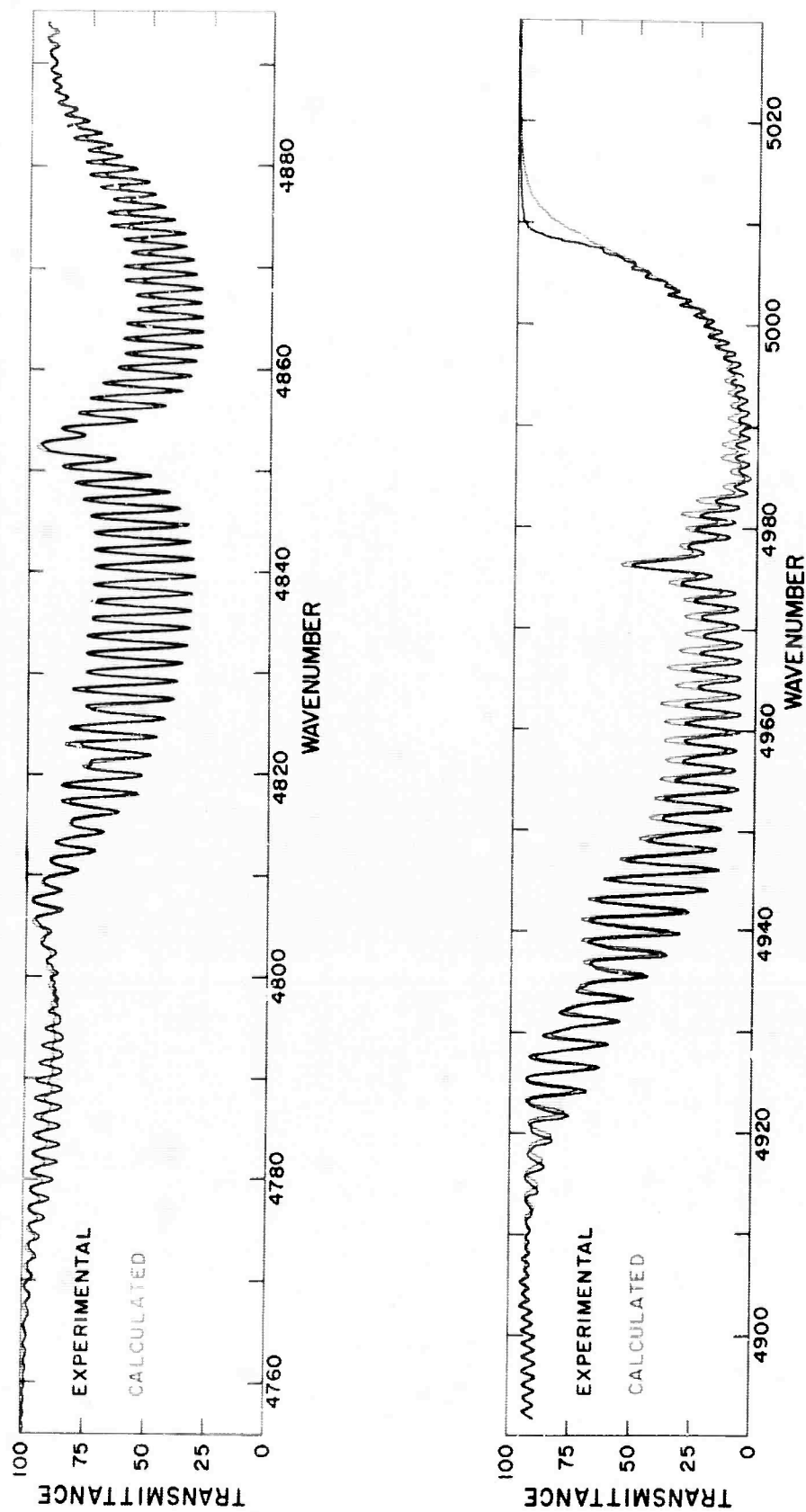
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Figure 2. The 2.0 micron bands of carbon dioxide at 1.0 atmosphere equivalent pressure and with 330 atmosphere cm path.
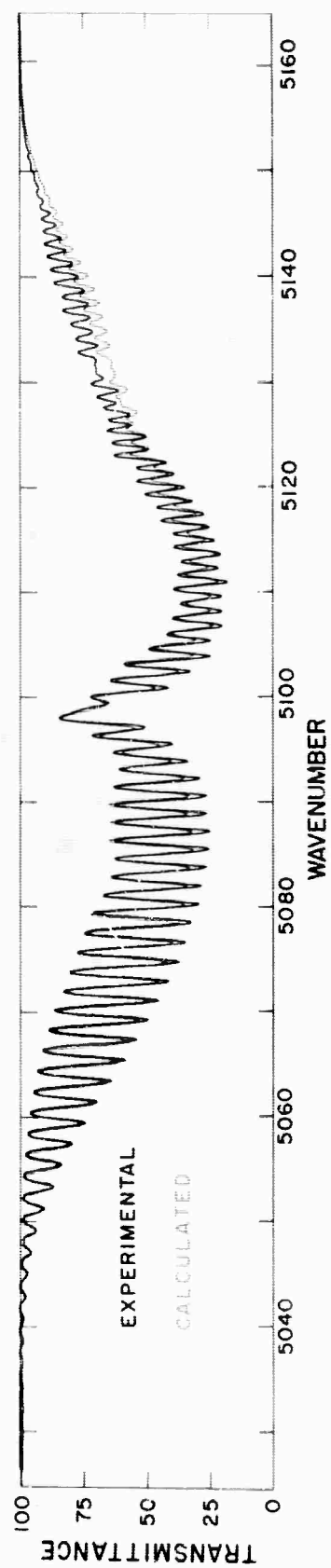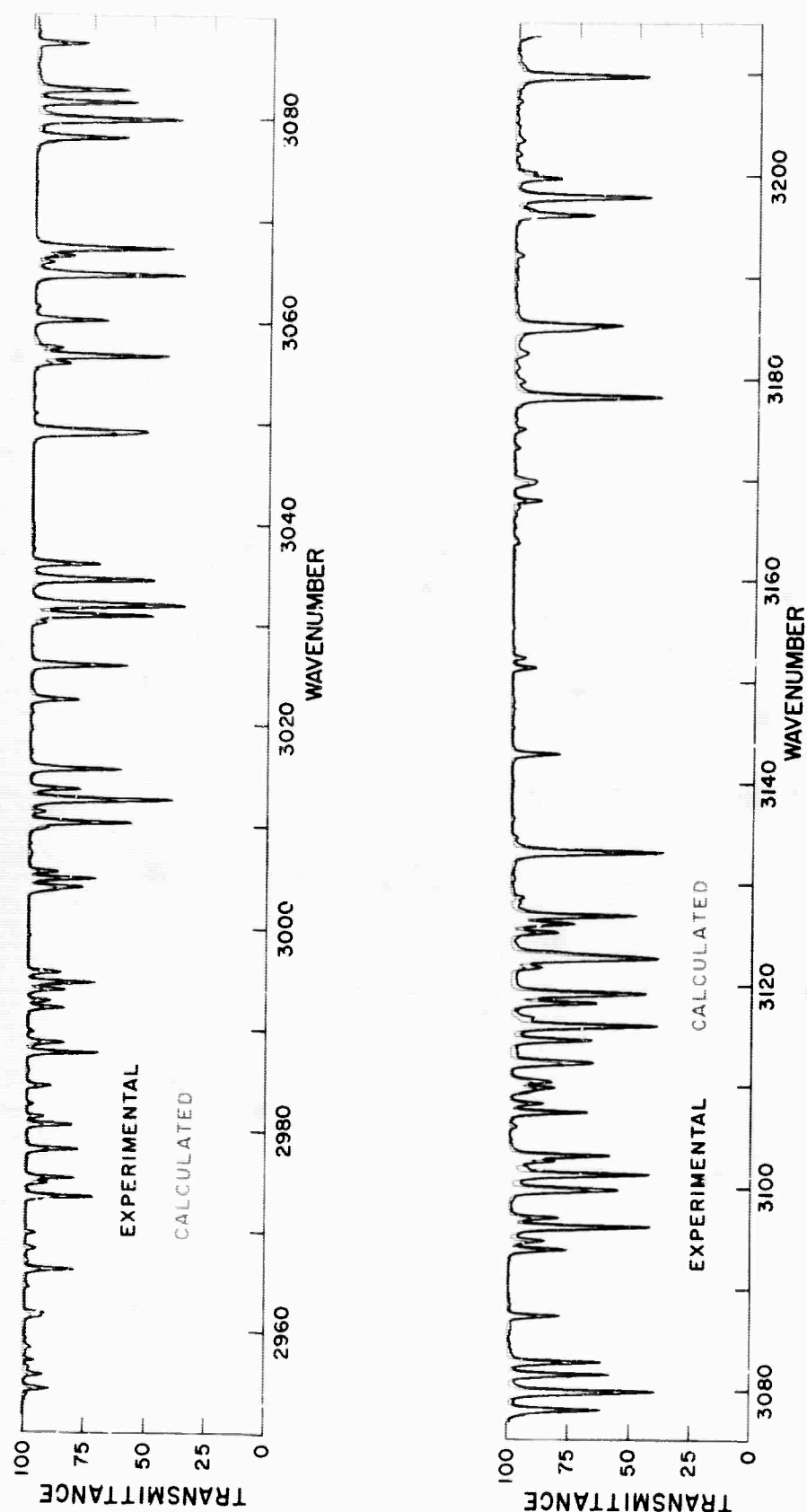
Figure 2. (Cont.)

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Figure 3. Water vapor spectra corresponding to a pressure of 0.332 atmosphere and an optical path of 0.0180 gm/cm^2.
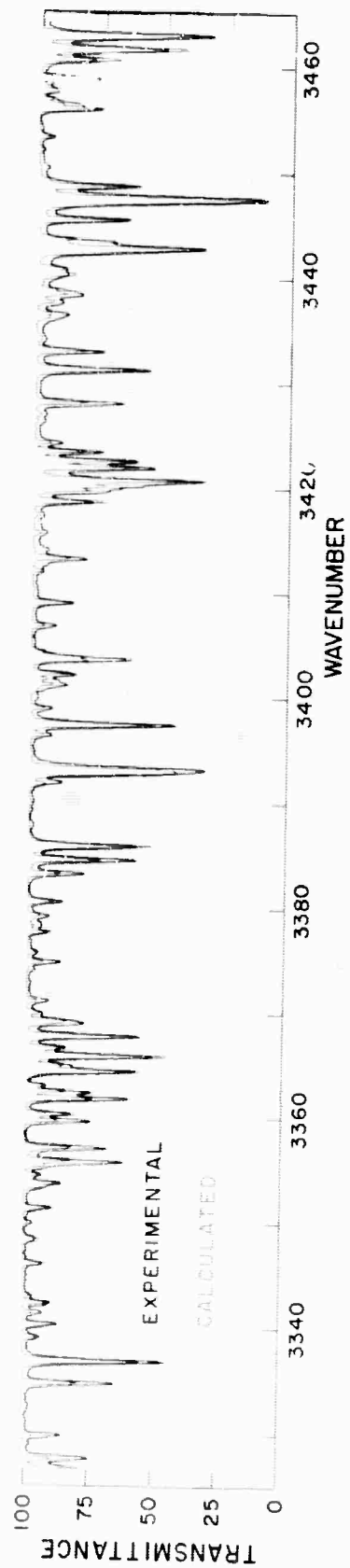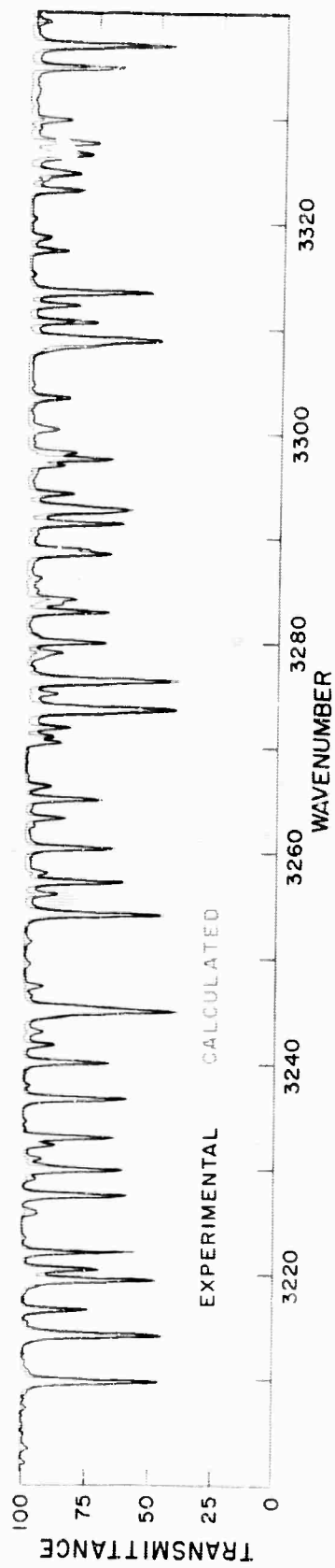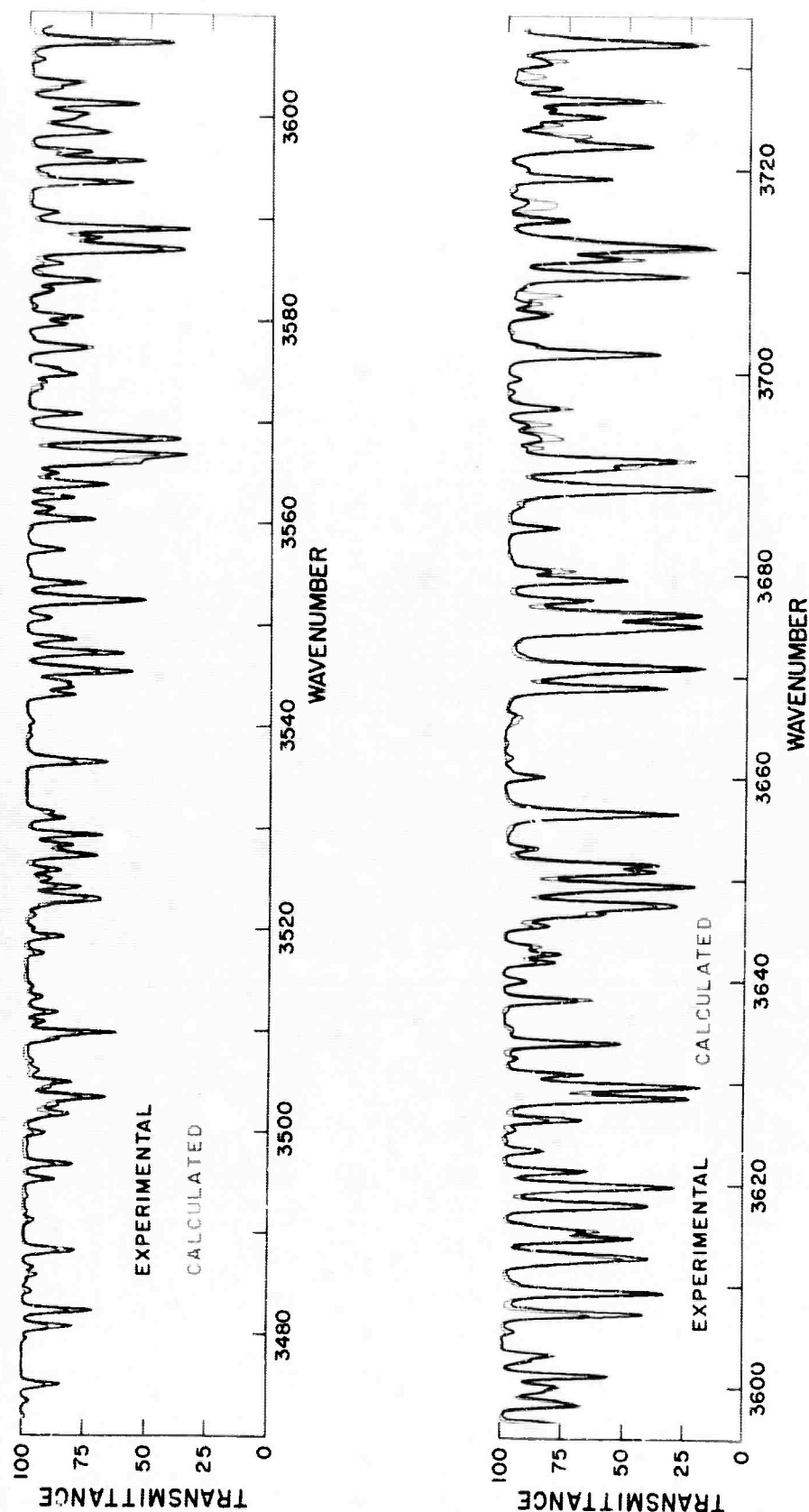
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Figure 3. (Cont.)

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Figure 4. Water vapor spectra corresponding to a pressure of 0.199 atmosphere and an optical path of 0.00169 gm/cm².
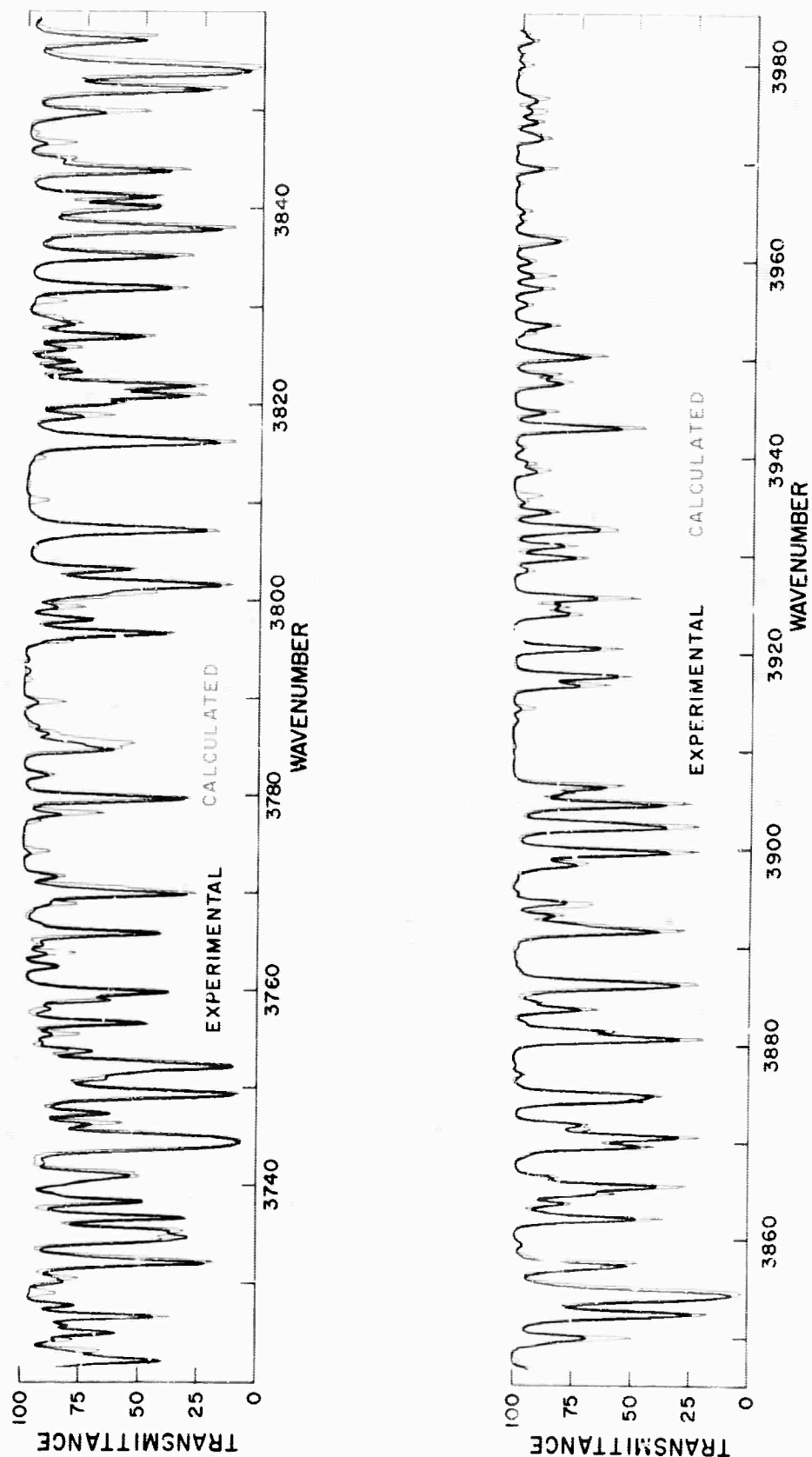
Figure 4. (Cont.)

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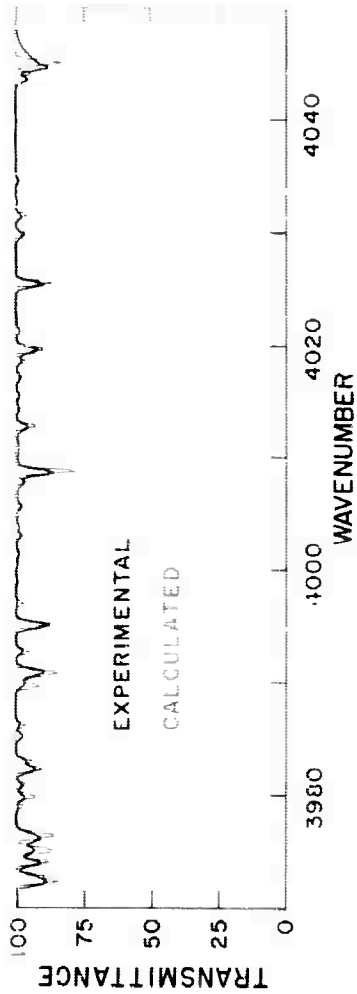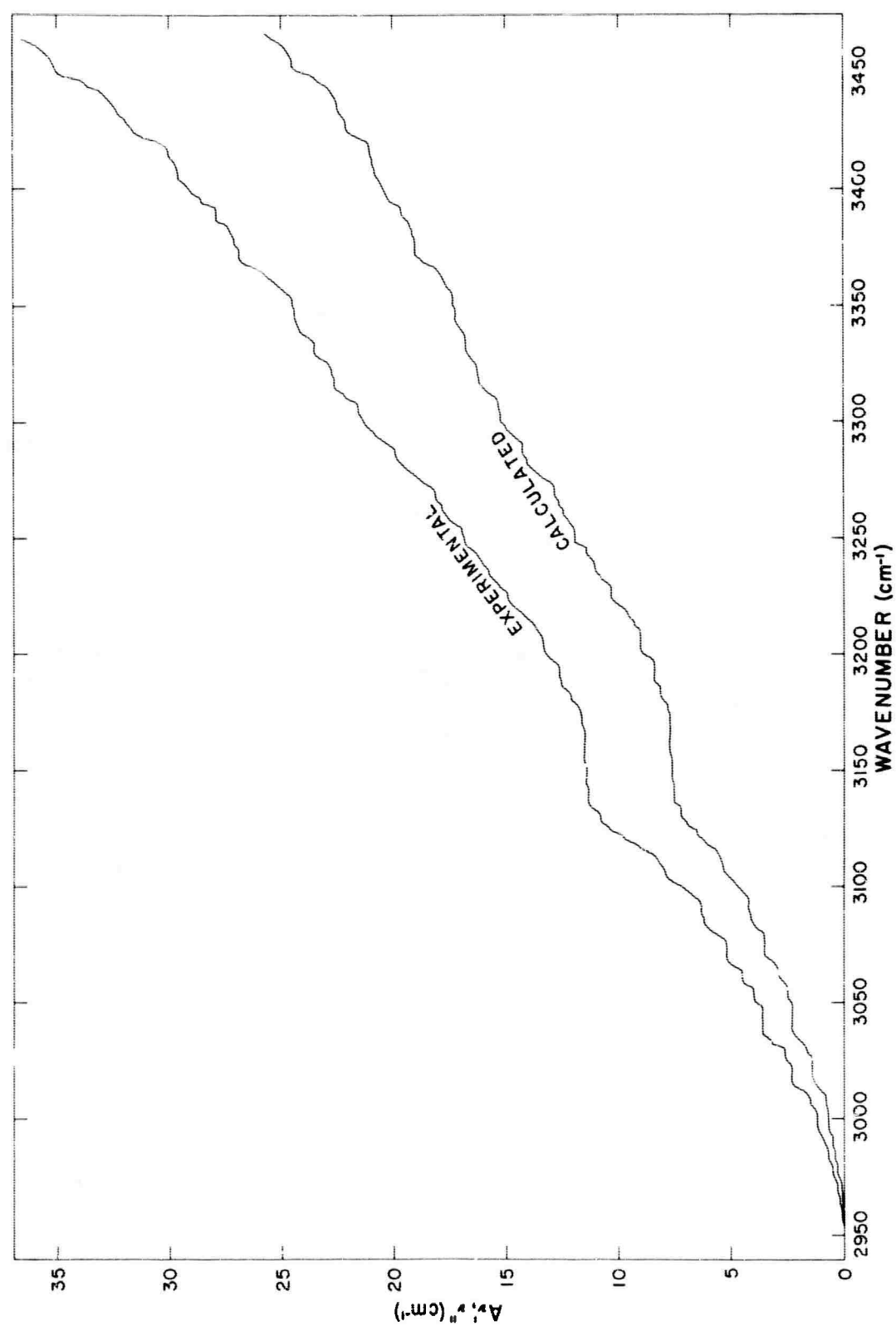Figure 4. (Cont.)

Figure 5. The integrated absorptance as a function of the upper frequency limit of the integral for the spectra shown in Fig. 3.
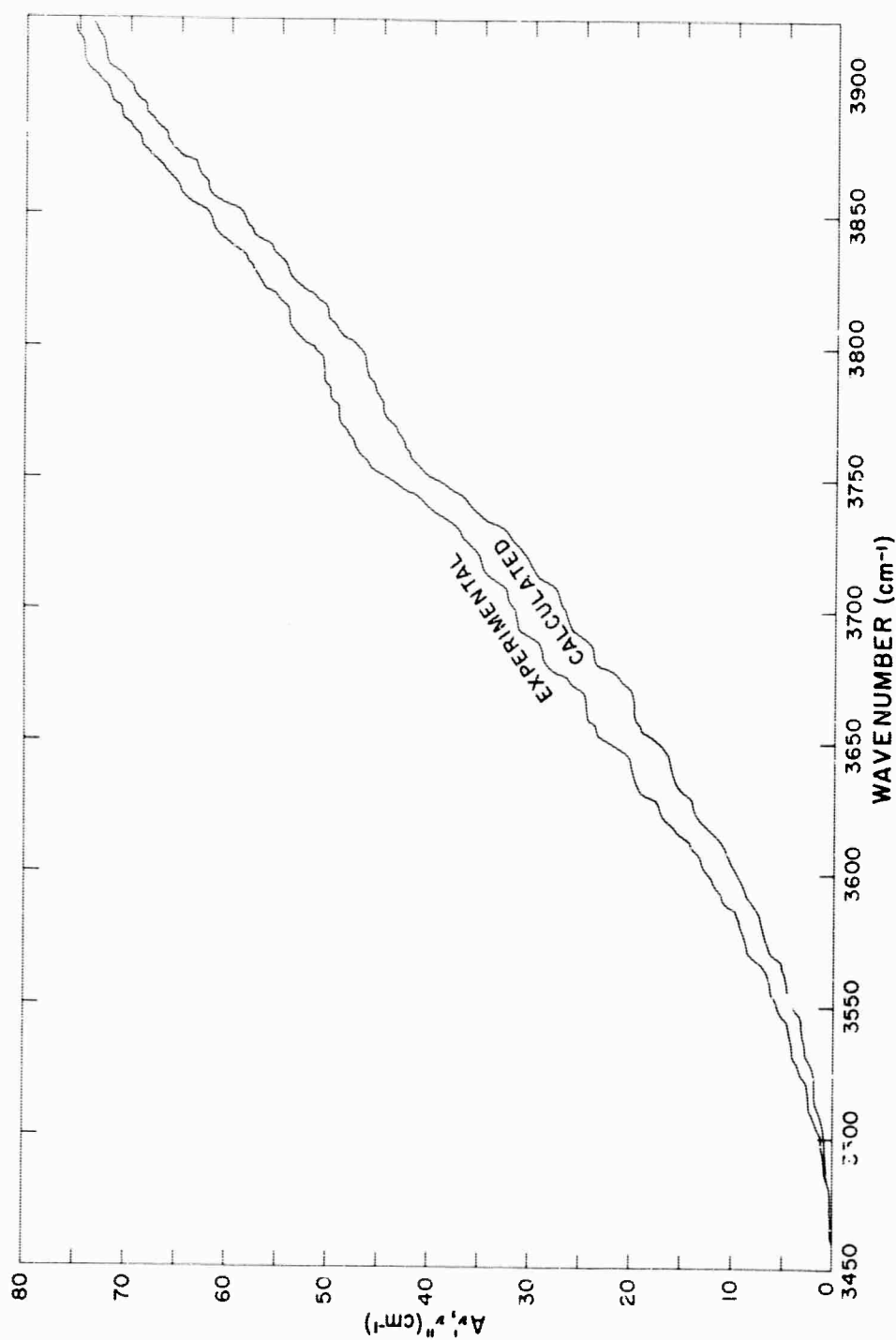
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Figure 6. The integrated absorptance as a function of the upper frequency limit of the integral for the spectra shown in Fig. 4.
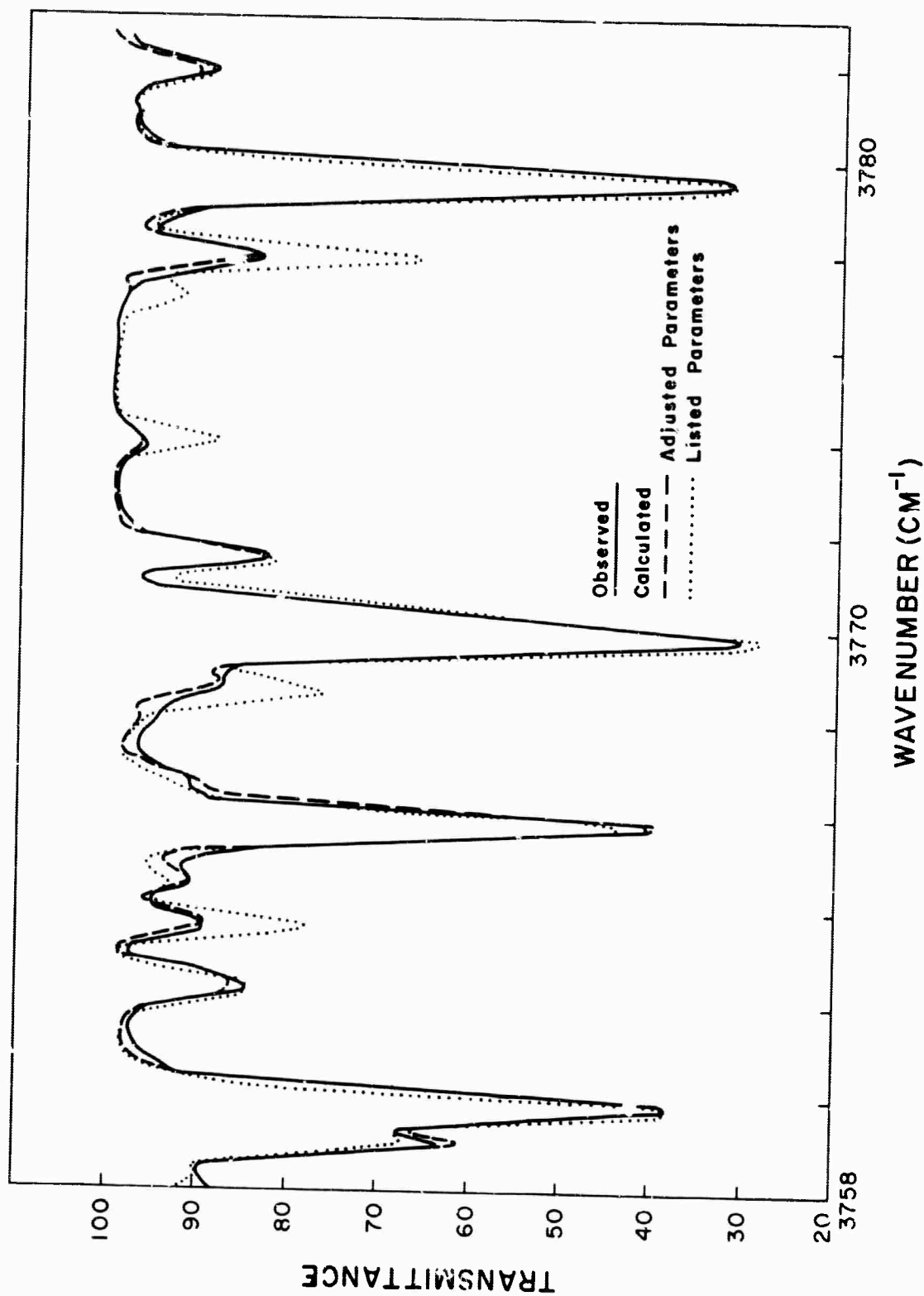
Figure 7. A portion of a measured water vapor spectra and calculated spectra using two sets of line parameters.

TABLE I

Interval	Path Atmos. Cm	Equivalent Pressure mm Hg	Integrated Absorptance cm^{-1}		Half-width at Atmosphere
			Experimental	Calculated	
1	50.33	77.52	67.02	68.05	0.07
1	50.33	77.52	67.02	70.93	0.08
1	50.29	762.05	104.23	108.00	0.08
1	16.91	762.05	78.77	84.62	0.08
1	0.3047	817.30	7.16	9.07	0.07
1	0.3047	817.30	7.16	9.34	0.08
2	50.33	77.52	63.70	61.73	0.07
2	50.33	77.52	63.70	64.03	0.08
2	50.29	762.05	88.42	91.17	0.08
2	16.91	762.05	74.22	75.43	0.08
2	0.3047	817.30	10.38	10.45	0.07
2	0.3047	817.30	10.38	10.83	0.08

Interval 1: 3490.0 - 3650.8 cm^{-1}

Interval 2: 3650.8 - 3766.0 cm^{-1}

TABLE II

Listed Values[5]		Adjusted Values	
Frequency	Strength	Frequency	Strength
3757.63	2.05	3757.61	-
3758.13	16.94	3758.05	34.94
3758.40	18.21	-	15.21
3758.63	7.36	-	-
3759.08	295.15	3759.01	-
3759.79	1953.34	-	-
3760.37	7.58	-	-
3761.51	1.28	-	-
3761.64	1.61	-	-
3762.18	43.30	-	-
3762.47	44.93	3762.52	-
3763.69	152.52	-	45.92
3764.16	3.15	-	-
3764.48	2.46	-	-
3764.58	17.29	3764.63	31.30
3765.79	1594.53	-	1902.14
3766.09	6.50	-	-
3766.39	14.26	-	16.26
3766.71	10.95	3766.76	16.96
3768.01	7.82	-	-
3768.66	142.82	3768.76	25.82
3768.93	12.74	3768.85	-
3769.52	2.36	-	-
3769.80	2641.47	3769.75	2201.47
3769.95	47.20	-	45.20
3770.15	37.88	-	-
3770.21	1.15	-	-
3770.48	433.22	3770.38	325.52
3771.45	78.28	3771.55	62.28
3771.68	26.13	3771.62	21.13
3773.97	2.84	3773.95	2.89
3774.05	49.24	3773.98	9.08
3777.11	26.15	-	3.15
3777.97	367.39	-	80.99

ACKNOWLEDGMENT

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Water Vapor						
Line Parameters						
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